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THERMOPHORESIS AND ITS THERMAL PARAMETERS FOR AEROSOL COLLECTION

Zhuo Huang, Michael Apte, and Lara Gundel

ABSTRACT

The particle collection efficiency of a prototype environmental tobacco smoke (ETS) sampler based on the use of thermophoresis is determined by optimizing the operational voltage that determines its thermal gradient. This sampler's heating element was made of three sets of thermophoretic (TP) wires 25μm in diameter suspended across a channel cut in a printed circuit board and mounted with collection surfaces on both sides. The separation between the heating element and the room temperature collection surface was determined in a numerical simulation based on the Brock-Talbot model. Other thermal parameters of this TP ETS sampler were predicted by the Brock-Talbot model for TP deposition. From the normalized results the optimal collection ratio was expressed in terms of operational voltage and filter mass. Prior to the Brock-Talbot model simulation for this sampler, 1.0V was used arbitrarily. The operational voltage was raised to 3.0V, and the collection efficiency was increased by a factor of five for both theory and experiment.

INTRODUCTION

Particulate matter (PM) is comprised of both solid and liquid particles that are suspended by gas molecules in the air. The composite mix of PM and the air in which it is suspended is defined as an aerosol. According to the U.S. Environmental Protection Agency (EPA), PM-2.5 and PM-10, which are particles with aerodynamic diameters less than 2.5 and 10μm, respectively, can cause adverse health effects with even short-term exposure. Environmental tobacco smoke (ETS) exposure increases the risk for respiratory diseases and lung cancer. The involuntary exposure for children (through secondhand smoke) causes lower respiratory illness in their infancy and early childhood and adverse effects on lung function across childhood [1]. Given the important public health implications of PM exposure, a continued need exists for development of cost effective, accurate, and practical measurement devices [2]. Various prototype instruments are being developed to measure the mass, size, and chemical composition of PM. Development of these devices is motivated by the desire to obtain a better exposure assessment tool to remedy misclassification of ETS exposure in epidemiological studies (see [1] Chapter 1 for details). The device under investigation measures particle phase (ETS) and gas phase (nicotine and 3ethenyl pyridine) ETS exposure. This portion of the project was focused on the ETS deposition mechanism of the particle phase sampling device.

The use of van der Waals induced dipole-dipole attraction is sufficient for surface retention of micron and sub-micron sized particles. Thermophoresis, the normal force on a particle created by the momentum transfer along the decreasing direction of a thermal gradient [3], is an effective method for particle collection in validating traditional methods [4]. When an aerosol is flowing in a narrow channel, the thermal gradient strength determines the strength of thermophoretic (TP) forces on the particles.

Experiments were conducted with a new device developed to sample airborne particles in indoor air with support from the California Tobacco Related Disease Research Program (TRDRP). Seeking a better understanding of thermophoresis to improve the collection efficiency became a motivation for exploring the theories dealing with TP particle deposition in laminar flow regimes in ducts (~500μm high). In a series of particle phase experiments using a TP ETS sampler, the TP voltage that controls the thermal gradient in the sampler was increased until the optimal (maximal) ETS deposition was determined.

The Brock-Talbot model for TP deposition [5] was previously used for proof of concept. In this project, this model was extended over a larger particle size range. A second purpose was to compare theoretical predictions with the experimental performance of the TP ETS sampler. The sampler used TP wires (California Fine Wire Co., Grover Beach, CA, Nickel Alloy 120) 25μm in diameter to heat up the surrounding air by applying a voltage across the wires. An arbitrary operational voltage of 1.0V was selected for powering the TP wire prior to computer simulation and chamber experiments. Since the temperature of the wires was proportional to the applied voltage, establishing the temperature-voltage relationship was necessary to obtain optimal collection.

MATERIALS AND METHODS

Theoretical Model

In the presence of a thermal gradient, the phenomenon of thermophoresis becomes significant in influencing particle motion [6]. TP deposition is based on a temperature difference between two close surfaces; given the appropriate gradient and particle size range, the collection can be 100% efficient [3]. The TP force is very effective for depositing tiny particles flowing through a channel onto its cooler surface. Other than the temperature gradient, many other parameters such as thermal conductivity, dimension of the flow channel, and viscosity associated with the particle stream play important roles.

The Brock-Talbot TP force model given by He and Ahmadi [5] is:

$$F_{th} = 2\,C_s\,C_c\,\frac{1}{\tau}\,\nu\,H\,\frac{\nabla T}{T}\,m, \qquad (1)$$

where C_s is the thermal slip coefficient, and C_c is the Stokes-Cunningham slip correction:

$$C_C = 1+\frac{2\lambda}{d}\left(1.257+0.4\,\mathrm{Exp}\left[-1.1\,\frac{d}{2\lambda}\right]\right), \qquad (2)$$

and τ is the relaxation time in equation (1) given by:

$$\tau = \frac{S d^2 C_C}{18\,\nu}. \qquad (3)$$

S is the ratio of particle density to fluid density [7]. T and ∇T are the room temperature and thermal gradient; m is the unit mass and v is the kinematic viscosity of the molecules. H is the molecular accommodation coefficient given by:

$$H = \left(\frac{1}{1+3C_m\,Kn}\right)\left(\frac{\frac{k_a}{k_p}+C_t\,Kn}{1+2\,\frac{k_a}{k_p}+2\,C_t\,Kn}\right). \qquad (4)$$

This equation indicates the relationship between particle diameter and thermal conductivity. Talbot found the values of the coefficients C_s=1.17, C_t=2.18, and C_m=1.14. C_t is a number of order unity which relates to temperature jump [5], and C_m is the momentum exchange coefficient that depends on the particle-gas configuration, and [5, 7]. Kn=2 λ/d is the Knudsen number, where λ is the mean free path of air and d is particle diameter; k_a and k_p are the thermal conductivity of gas (air) and particle respectively.

The TP collection ratio:

$$\eta = \frac{U L}{2\,h\,u_m} \qquad (5)$$

is the ratio of the TP velocity and the mean air velocity u_m of the channel. Here, L and h are the length and the height of the channel. They are chosen to be 71.0mm and 0.5mm for the simulation, reflecting the dimensions of the TP ETS sampler. U is the TP velocity:

$$U = \frac{F_{th}}{m}\,\tau \qquad (6)$$

which, in effect, determines the TP collection ratio. Substituting τ and H into Equation (1) and expanding in terms of its dependent parameters, the equation becomes:

$$F_{th} = 36\,C_s\,\frac{\nu^2}{Sd^2}\left(\frac{1}{1+3\,C_m\,Kn}\right)\left(\frac{\frac{k_a}{k_p}+C_t\,Kn}{1+2\,\frac{k_a}{k_p}+2\,C_t\,Kn}\right)\frac{\nabla T}{T}\,m \qquad (7)$$

The C's are collectively referred to as the drag forces. One can see that the TP force strength is inversely dependent on particle diameter and the temperature of forming the thermal gradient, where *v* and k_a/k_p dependence are accounted for by the type of particles. This equation is simulated with the molecular diameter of air chosen to be 5.7nm, and the aerosol particles consist of NaCl (for consistency with the reported work in [5]) with thermal conductivity k_p=6.69 $W\,m^{-1}\,K^{-1}$.

Apparatus

The collection apparatus consists of a loom-like frame machined from a copper clad printed circuit board that holds a set of TP wires and acts as a flow channel, and a mounting surface for fin-cooled aluminum particle collection surfaces. When assembled, the device becomes an air-tight flow-through channel with three sets of TP collection areas; a small pump of flow rate 0.01L/min was used to draw a PM-laden air sample through the device. Four fine TP wires of 25μm in diameter about 5mm in length were soldered, physically in parallel, and electrically in series to form a coplanar resistive heater about 5mm on a side on the collector frame. Three of these heaters were assembled on a single sampler to create three separate TP collection regions (see Figure 1). When the TP ETS sampler was assembled and a voltage was applied across one of the three wire

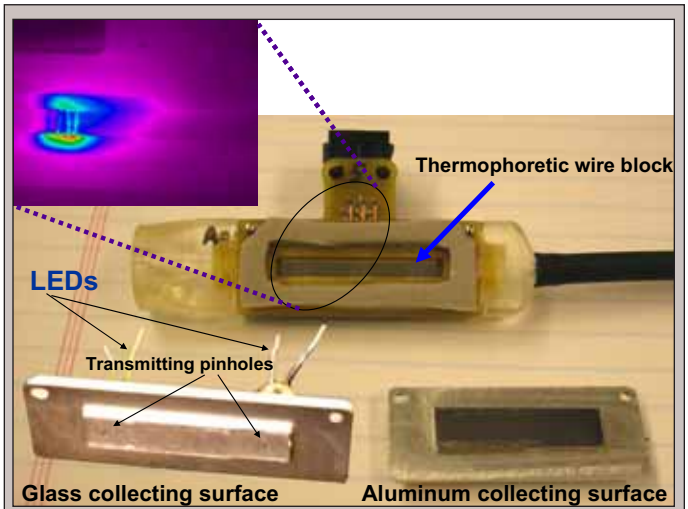

Figure 1. The TP ETS Sampler with thermophoretic wires indicated; glass collecting surface and aluminum collecting surface are separately shown at the left and the right. The thermogram insert shows the heated TP wires region when powered.

circuits, a thermal gradient was formed between the wires and the fin-cooled aluminum collection surface (Figure 1).

Two collecting surface configurations were used on the two sides of the TP wire blocks for particle collection. An aluminum collection surface (Figure 1) was fabricated with a polished thin aluminum surface attached to an aluminum body with double-sided heat conducting tape. A glass collection surface (opposite) was assembled with a thin glass cover slide and another aluminum body. The glass cover slide was glued at its edges to allow light transmission through two circular pinholes of diameter ~ 1mm that were each centered on a TP particle deposition area on the surface. LEDs (Roithner Lasertechnik, Vienna, Austria, UV-LED 380D30 and ELD-810-525) of peak wavelength of 380 nm (UV) and 810 nm (IR) were inserted into the back of the aluminum body allowing a light beam to be transmitted through the pinholes and to emerge normal to the glass surface. The LED pinholes were located in the center of the TP deposition surfaces for two of the three collection areas on the glass-covered collection body.

Thermophoretic Wire Temperature

In Figure 1, the thermogram insert shows the heated TP wires. Due to the low emissivity of the wires, the thermogram merely indicates the wires were hotter than their surroundings when powered, yet does not accurately indicate the *exact* temperature. Using an alternate approach to thermography, the approximate temperature of the TP wire could be calculated in terms of resistance with the known temperature coefficient of resistance of the TP wire provided by the manufacturer. A 50 x 50 x 80cm convection oven was used to measure the temperature-resistance relationship of the TP wires. A differential temperature controller gradually raised the temperature while two small 12V fans were used for air mixing inside the oven. A sampler was placed inside the oven and two blocks of TP wires' resistance were measured for comparison. The resultant voltage vs. temperature relationship could be obtained for determining the associated temperature with experimental voltage and current using Ohm's law.

Experiment Setup and ETS Generation

A 24m^3 environmental chamber was built with a wooden frame, wallboard walls and ceiling, and magnetic refrigerator door seals. Low volatile organic compound emitting paints and sealants were applied inside the chamber to minimize unwanted room-ambient particles and volatile organic compounds [8]. The chamber was bolted in place in a one-story building, no ventilation equipment was used, and the room temperature stayed approximately steady during the sampling periods. The TP ETS sampler was hung from a stand 0.5 meters above the floor, side by side with the a Personal Environmental Monitor (PEM) PM-2.5 sampler (SKC Inc., Eighty Four, PA, 761-203). The PEM, assembled with a Teflon-coated fiberglass filter (SKC Inc., Eighty Four, PA, SKC Filter 761-203) and sampling at a rate of 2.0L/min, provided the integrated PM mass concentration in the chamber during the sampling period. Using the proportionality of the integrated PM mass concentration and TP ETS sampler flow rates, the total particle mass that traversed the TP ETS sampler during operation was calculated.

ETS was generated by concurrently and completely smoldering 8 cigarettes for each run. ETS is a complex mixture of solid and vapor phase particles produced primarily from the burning of cigarettes and is dependent on the puff rate [8]. The TP ETS sampler validation was focused on the deposition mechanism, thus smoldering cigarette smoke was effective and appropriate for these experiments. The TP ETS sampler controller microprocessor was programmed to switch amongst the 3 sampler TP wire regions in a reversing sequence fashion (i.e., 1-2-3, 3-2-1, 1-2-3,…) every 5 minutes. The purpose of this was to avoid collection bias during decay of the aerosol concentration in the chamber, and therefore this non-random sampling pattern distributed near-equal particle mass collection on each of the three regions of the sampler's collection surfaces. A series of experiments were conducted at increasing wire voltage settings from the pre-set voltage of 1.0V to the optimal voltage of 3.0 volts where a maximum particle collection value (see Table 1) was reached. The voltage and current inputs of each wire were recorded for analysis. Each experiment was allowed to collect for at least 17 hours in order to obtain significant amount of ETS.

Spectrometry and Related Statistics

Thermal Conductivity Ratio (k_p/k_a) of Several Materials

TP Voltage (V)	Resistance (Ω)	T [°C]	∇T [K/cm]
1.0	13.6	107	1740
1.5	15.4	179	5190
2.0	18.3	252	4640
2.5	19.5	325	6090
3.0	25.2	397	7540

Table 1. The corresponding thermal parameters for the TP voltages used.

The samples on the glass slide collection surface were then examined by using fiber optic probes connected to a UV-NIR spectrometer (Ocean Optics, Dunedin, FL, S2000 UV-VIS 250800nm). Figure 2 shows the schematic of the operation when the UV LED was powered. The LEDs were placed on a light-tight housing and connected to an external power supply. With the LEDs turned on, UV or NIR light was transmitted through the cover slide. Fiber optic probes were attached to the bottom of the housing cavity and connected via a channel selector to the spectrometer. After ETS was collected on the glass cover slide the sample spectrum was recorded. Next, the ETS sample was removed by cleaning off the glass collection surface to obtain the baseline spectrum. The absorbance was calculated with Beer's Law, $A = -\log(I/I_0)$, where I and I_0 were the calculated peak intensities of the spectrum taken by a spectrometer.

To account for small differences between ETS concentrations in each experiment, the calculated ETS absorbance was normalized by the integrated ETS concentration from the PEM during the sampling period in order to determine collection efficiency in

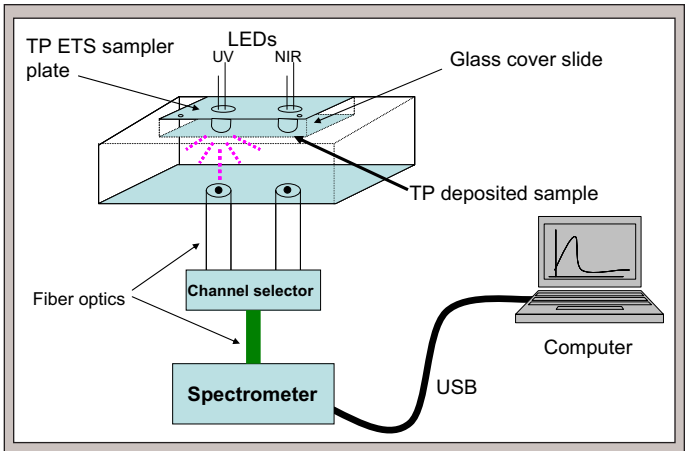

Figure 2. Configuration of spectrometric method for measurement of particle deposition on TP ETS Sampler.

Thermal Conductivity Ratio (k_p/k_a) of Several Materials	
Materials	Unitless k_p/k_a
Air at 20°C	1
Asbestos	3
Paraffin oil	5
Magnesium oxide	5
Stearic acid	5
Castor oil	7
Water	23
Clay	27
Glass	32
Fused silica	38
Granite	81
Carbon	169
Sodium chloride	254
Mercury	323
Quartz	369
Iron	2573

Table 2. Ratio of thermal conductivity of several materials to air. kp and ka values were adopted from Ref. 3.

terms of mass. The UV absorbance results were much more robust than those from the NIR LED since ETS has strongly enhanced UV absorbance. The NIR absorbance measurements in the case of these ETS samples were typically below the limit of quantitation for the method, making them of little use in this investigation. The filter mass collected by PEM Teflon-coated fiberglass filter was first measured with an electronic microbalance, and then the air PM concentration was calculated based on the sampling flow rate and duration. The ETS mass that entered the TP ETS sampler during each experiment was computed by multiplying the PEM-based PM concentration by the experiment sampling time and sampler flow rate. The TP ETS sampler UV absorbance was normalized to that of a 10μg mass sampling based on the above calculation.

RESULTS

Collection Ratio

The Brock-Talbot deposition ratio was plotted as a function of aerosol diameter for NaCl in Figure 3a. Figure 3b depicts the influence of the thermal conductivity of different materials on thermophoretic deposition. Table 2 lists the thermal conductivity ratio (kp/ka) of several possible material types that can be found in PM. These results taken together show how the different types of PM could have different thermal properties that affect the TP collection ratio. Figure 3a shows that the thermophoretic particle deposition ratio is about 0.6 and gradually decreases to 0.1 for the pre-set thermal gradient (∇T=1600K/cm), from 10nm to about 1 micrometer.

Temperature Correlation

Heating the TP wires inside the oven generated resistance vs. temperature data that matched well with the wire manufacturer's temperature coefficient of resistance (see the dashed line in Figure 4a). The voltage applied to the TP wire during sampling was recorded each time and the correlation to resistance was plotted (Figure 4b).

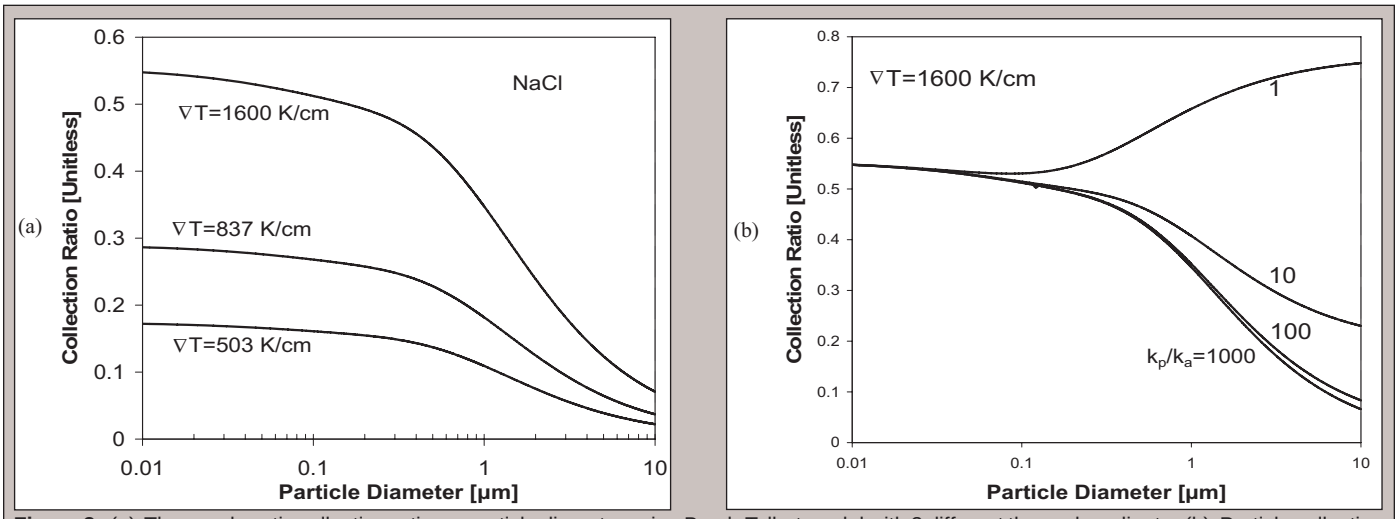

Figure 3. (a). Thermophoretic collection ratio vs. particle diameter using Brock-Talbot model with 3 different thermal gradients. (b). Particle collection ratio vs. diameter for different kp/ka using the Brock-Talbot model.

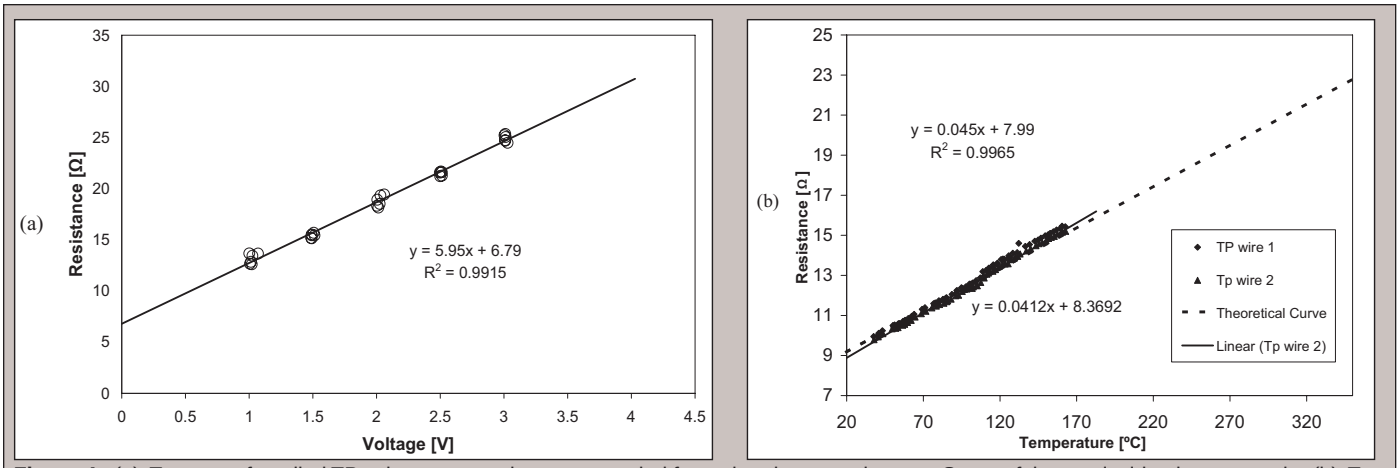

Figure 4. (a). Ten sets of applied TP voltages vs. resistance recorded from chamber experiments. Some of them coincide almost exactly. (b). Two different blocks of wire were heated and their resistances were measured. The dashed line is the curve calculated with manufacture resistance coefficient, α=0.0045, plotted for a wider range than the experimental range.

Thus, by comparing Figure 4a to the former graph one can obtain the temperature as a function of applied voltage. Combining the fitted equation from Figure 4b and the theoretical curve from 4a, a relationship could be simply described as T = 145.1V - 28.5, where T is the TP wire temperature (°C) forming the thermal gradient and V is the applied voltage. Table 1 summarizes the experimental applied voltages and their thermal parameters.

Normalized Absorbance and Optimal Collection

The chamber experiment data show increased particle collection in terms of absorbance with increasing wire voltage. ETS absorbance measurements from the TP ETS sampler normalized to a 10µg sample was plotted against TP wire voltage and fitted with a 2nd order polynomial (see Figure 5). Each data point was obtained by averaging two experiments. Error bars in Figure 5 represent the greater deviation from the experimental average. The optimal voltage for this ETS sampler was identified to be between 2.5 and 3.0V based on this fit.

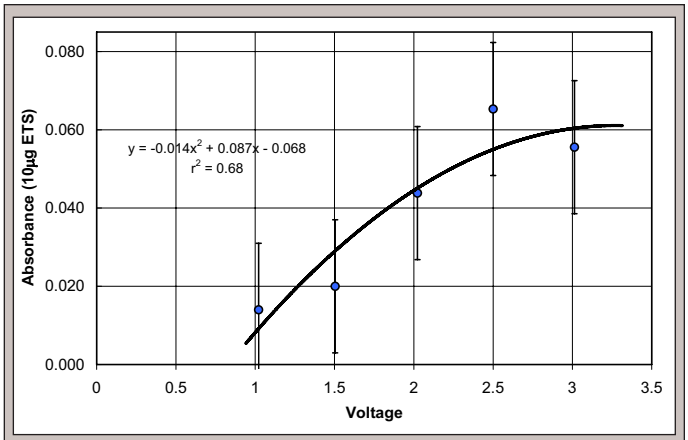

Figure 5. Particle phase titration result shows the optimal collection of the TP ETS sampler is with the input voltage of about 2.5V; the scattered absorbance data are averaged with two filter mass (PEM) normalized data. An enhancement by a factor of 5 is suggested. The error bars represent the greatest deviation from the experimental average based on two experiments for each voltage.

DISCUSSION AND CONCLUSIONS

Thermophoretic modeling (Figure 3a) shows an effective collection ratio for sub-micron particles. The collection ratio is defined as the ratio of TP velocity to the mean air velocity in the channel and the collection efficiency is the ratio of aerosol being collected to the amount that flows in the channel. Figure 3a also shows that the particle collection ratio quickly decreased in the micron region. As the diameter increases further (Kn<2), the collection ratio decreases drastically to 0.1. The Brock-Talbot simulation predicts that particle deposition will increase substantially as the thermal gradient is raised above T=1600K/cm, a value that was previously set for this device. If the ratio kp/ka is small, the diameter of the particle determines the value of Equation (3) as the Ct Kn term dominates; on the other hand, with kp/ka increasing the collection ratio decreases drastically as seen in Figure 3b. This result is consistent with the presentation by Hinds on the subject of thermophoretic velocity as a function of kp/ka [3].

Based on the Brock-Talbot theoretical prediction, we should expect higher particle deposition for sub-micron sized particles. The calculated thermal gradient for 2.5V is about ∇T=6090K/cm, and the Brock-Talbot model collection ratio for this thermal gradient is well above 1.0. The original selection for TP voltage of this low-cost particle sampler took into account the power consumption; 1.0V TP voltage was believed to be above 80% efficient for PM-2.5 collection according He and Ahmadi's results [5, Figure 6]. This study has shown, both numerically and experimentally, the optimal deposition ratio occurs between 2.5 and 3.0V for this TP ETS sampler. The absorbance at 2.5V TP collection shows a signal five times stronger than that of 1.0V (Figure 5). It is obvious that the optimal collection efficiency in term of normalized ETS mass is at least a factor of 5 higher than the 1V thermal gradient. Figure 6 also shows the dependence of deposition ratio on voltage, verifying the Brock-Talbot model. This also shows an enhancement by a factor

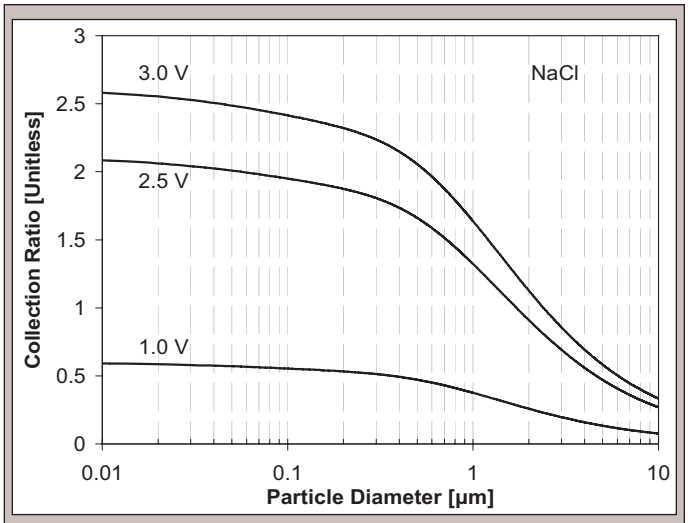

Figure 6. The Brock-Talbot model with thermal gradient corresponding to 1.0 ($\Delta T = 1740K/cm$), 2.5 ($\Delta T = 6090K/cm$), and 3.0V ($\Delta T = 7540K/cm$) applied voltage (TP gradient). Result shows that 3.0V applied voltage can collect a factor of 5 more than that of 1.0V applied voltage.

of 5 (from about 0.5 to 2.5 for a 100nm particle diameter). This result may provide a useful calibration mechanism for developing the TP ETS sampler.

Future Work

This project investigated several major issues dealing with the TP ETS sampler particle collection efficiency and identified some important thermal parameters that influence aerosol collection by thermophoresis. This result raised a concern that the oven-treated TP wire temperature may not correctly describe the thermal gradient of the TP ETS Sampler's in-situ performance. Calibrating the sampler's thermal gradient will be helpful because the configuration of TP wires relative to a cool flat collection surface differs from the two-parallel plate configuration used in the Brock-Talbot model [7, 9]. We have shown that the applied voltage for the TP wires should be higher. Moreover, how the TP wire temperature affects the thermal gradient is unsolved for the configuration of this sampler. This question can be answered with future investigation.

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